ISOMER SHIFT CALIBRATION OF ^{57}Fe BY LIFE-TIME VARIATIONS IN THE ELECTRON-CAPTURE DECAY OF ^{52}Fe.

J. Ladrière*, A. Meykens, R. Coussement, M. Cogneau*, M. Boge**, P. Auric**, R. Bouchez+, A. Benabed+ and J. Godard+

Instituut voor Kern- en Stralingsfysika, Universiteit Leuven, Belgium
* *Laboratoire de Chimie inorganique et nucléaire, Université de Louvain, Louvain-la-Neuve, Belgium*
** *D R F, Centre d'Etudes Nucléaires de Grenoble, France*
\+ *I S N, Université de Grenoble, France*

Résumé.- Des mesures de variation de probabilité de capture électronique du ^{52}Fe dans différents composés de fer permettent d'estimer les densités électroniques au noyau. On en déduit une constante de calibration α pour le déplacement isomérique du ^{57}Fe : $\alpha = -0{,}31 \pm 0{,}04\ a_0^3\ .mm.s^{-1}$.

Abstract.- Electron densities at the nucleus are estimated from measurements of the change in electron capture rate of ^{52}Fe in different iron compounds. This leads to a calibration constant α for the ^{57}Fe isomer shift : $\alpha = -0.31 \pm 0.04\ a_0^3\ .mm.s^{-1}$.

1. Introduction.- The Mössbauer isomer shift δ between a pair of chemical compounds is given by

$$\delta = \frac{2\pi Ze^2}{3} \Delta \langle r^2 \rangle . \Delta\rho = \alpha . \Delta\rho$$

where α is a constant depending on $\Delta\langle r^2\rangle$, the change in mean square charge radius of nucleus during the Mössbauer transition, and $\Delta\rho$ is the difference of the electron densities at the nucleus for the two chemical states.

The calibration of the isomer shift consists in the determination of α or $\Delta\langle r^2\rangle$ by combination of estimated $\Delta\rho$ values with the corresponding measured isomer shift.

In the present work, $\Delta\rho$ values are obtained from life-time measurements in iron compounds labeled with radioactive ^{52}Fe. This experiment uses the proportionality of the electron capture decay constant λ_{EC} to the electron density at the nucleus. For the electron capture (44%) in ^{52}Fe one has that $\Delta\lambda/(0.44\lambda) = \Delta\lambda/\lambda_{EC} = \kappa\Delta\rho/\rho$, where λ is the total decay constant and $\kappa = 1$. (This last factor could however be equal to 1.3 if exchange and overlap effects are taken into account during the EC /1/).

2. Experimental.- Direct measurements of $\Delta\lambda$ for several pairs of chemically different ^{52}Fe : iron compounds have been performed using differential ionization chambers (ionization balance). This instrument provides a relative accuracy less than 5×10^{-5} for $\Delta\lambda/\lambda$, as previously mentioned /2/, so that effects of the order of 10^{-4} can be readily measured.

In order to obtain the same total efficiency versus time for the two chambers, the counting geometry (4π), the initial activity (1 ± 10^{-3} mCi) and the self absorption coefficients of both sources are carefully adjusted. This last adjustment is realized by adding (except for one experiment) to each labeled compound the appropriate amount of inactive paired compound. Furthermore, for each pair of sources, two measurements are made by interchanging the sources from one chamber to the other, in order to cancel the zero effect of the apparatus in the arithmetic mean of reverse results.

Since the largest $\Delta\lambda$ values are expected for the largest isomer shift differences $\Delta\delta$, the following iron compounds have been synthetized and paired as indicated in table I (δ values at 2.90 K).

Compound	δ_{Fe} (mm.s^{-1})
(O): $FeC_2O_4.2H_2O$	+ 1.19 ± 0.02
(H): α-Fe_2O_3	+ 0.38 ± 0.01
(G): $KFe(CH_2OCO_2)_2H_2O$	+ 0.40 ± 0.01
(C): $K_4Fe(CN)_6.3H_2O$	- 0.05 ± 0.01
(F): $BaFeO_4$	- 0.90 ± 0.02

The different sources used are tested by Mössbauer spectroscopy after the decay of ^{52}Fe.

The ^{52}Fe ($t_{1/2}$ = 8.3 h.) activity is produced by a $^{55}Mn(p,4n)^{52}Fe$ reaction at a proton energy of 65 MeV, and separated from the Mn activities present by anion exchange. Gamma-ray spectroscopy of the sources five days after their production, i.e. after the differential measurements, checks the selectivity of the separation. Only very small amounts of ^{54}Mn are found in about the same proportion in

both sources.

Table I

Measured relative differences of the electron-capture rate and the deduced differences between the corresponding electron densities.

$^{*}\delta_{-(a)} - {}^{*}\delta_{+(a)}$	Source combination		$\frac{\lambda_{-(a)} - \lambda_{+(a)}}{\lambda} \times 10^{4}$	$\frac{\Delta\lambda}{\lambda} \times 10^{4}_{(b)}$	$\Delta\rho_{(b)}$
$(mm.s^{-1})$	+(a)	-(a)			(a_o^{-3})
0.00	*G	*G	+0.20 ± 0.19	+0.18 ± 0.18	-0.62 ± 0.62
0.00	*O	*O	+0.17 ± 0.18		
-0.81	*O.H	*H.O	+0.89 ± 0.20	-0.77 ± 0.22	-2.64 ± 0.75
+0.81	*H.O	*O.H	-0.64 ± 0.39		
-0.81	*O	*H	-0.46 ± 0.19	-0.97 ± 0.20	-3.32 ± 0.68
+0.81	*H	*O	-2.40 ± 0.36		
-1.24	*O.C	*C.O	+1.29 ± 0.15	-0.98 ± 0.14	-3.34 ± 0.48
+1.24	*C.O	*O.C	-0.67 ± 0.23		
-2.09	*O.F	*F.O	+1.29 ± 0.48	-1.93 ± 0.33	-6.61 ± 1.13
+2.09	*F.O	*O.F	-2.56 ± 0.45		

(*) refer to the ^{52}Fe labeled compound.
(a) refer to the sign of the collected current.
(b) arithmetic mean value of symmetric experiments for $\Delta\delta$ positive.

Because of its life-time ($t_{1/2}$ = 303 days) this ^{54}Mn does not perturb the measurements. Nevertheless, it is important that the synthesis of the compounds to be compared does not modify the ^{52}Fe-^{54}Mn ratio.

3. Results and discussion.- The results are summarized in table I. Errors are pure statistical. The relative change in the decay constant is the fitted $(\lambda_- - \lambda_+)$ value, divided by the literature value of $\lambda = (2.327 \pm 0.002) 10^{-5}.s^{-1}$ /3/. From these values, it is seen that no important asymmetry of the two chambers appears while reversing the sources; the sign of $(\lambda_- - \lambda_+)$ reverses too. For one experiment, however, it is not the case, because the self absorption of the two sources are different and, as a result, the zero of the apparatus is shifted. Fortunately, the arithmetic mean of reverse results cancels this zero.effect, within the statistical error.

Figure 1 shows the linear least square fit of the experimental $\frac{\Delta\lambda}{\lambda}$ values versus the isomer shift. Using the relativistic value of $\rho = 15070\ a_0^{-3}$ /4/, where a_0 is the Bohr radius, the calibration constant α and $\Delta\langle r^2\rangle$ can be deduced from the slope of that linear relationship :

$\alpha = - 0.31 \pm 0.04\ a_0^3.mm.s^{-1}$ and

$\Delta\langle r^2\rangle = - (28 \pm 4)\ 10^{-3} fm^2$

(or $\alpha = - 0.40 \pm 0.05\ a_0^3.mm.s^{-1}$ and $\Delta\langle r^2\rangle = - (37 \pm 5)\ 10^{-3} fm^2$ if the factor k is taken equal to 1.3).

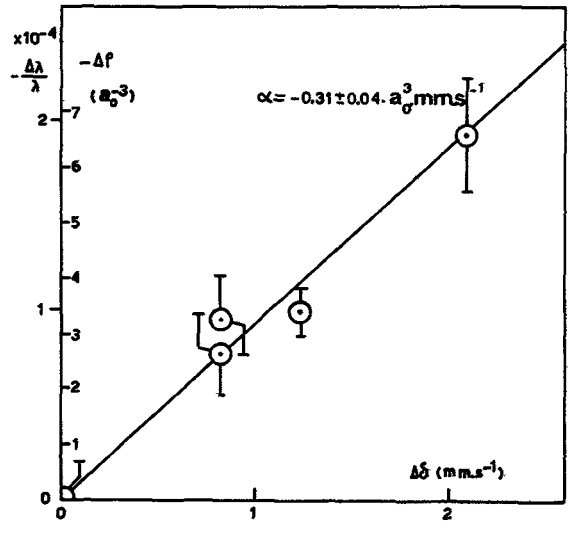

Fig. 1 : Electron density differences versus the corresponding isomer shift. (The left hand scale gives the corresponding measured relative changes of decay rate).

In both cases, this result is surprinsingly different from the experimental value of Rüesgsegger and Kündig /5/.

Acknowledgements.- The authors wish to thank the staff of the cyclotrons of Louvain-la-Neuve and Grenoble for the production of the ^{52}Fe activity. The financial support of the "Interuniversitair Instituut voor Kernwetenschappen" is greatly acknowledged.

References

/1/ Bahcall, J.N., Phys. Rev. 132 (1963) 362.

/2/ Boge, M., Meykens, A., Revue Phys. Appl. 11 (1976) 167.

/3/ Rothman, S.J., Peterson, N.L., Chen, W.K., Hines, J.J., Bastar, R., Robinson, L.C., Nowicki, L.J., Anderson, J.B., Phys. Rev. C9 (1974) 2272.

/4/ Mallow, J.V., Freeman, A.T., Desclaux, J.P., Phys. Rev. B13 (1976) 1884.

/5/ Ruegsegger, P., Kündig, W., Phys. Lett. 39B (1972) 620 and Helv. Phys. Acta 46 (1973) 165.